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6. AUTHOR(S) Robert W. Boyd				
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13. ABSTRACT (Maximum 200 words) Laboratory research has been performed to assess new materials for use in nonlinear optics. One aspect of this work involves the formation of composite materials. This work is motivated by recent theoretical predictions that composite materials can possess a nonlinear susceptibility exceeding those of its constituent materials. We verified this prediction by constructing a composite material formed of alternating layers of titanium dioxide and the conjugated polymer poly paraphenylene benzobisthiazole, and demonstrating that the composite possessed a third-order susceptibility 35% larger than that of the polymer, its more nonlinear constituent. A second aspect of this work involved measuring the third order susceptibility of a family of related polymers. The results of these measurements provide insights into the relationship between the structure of polymers and the size of their optical nonlinearities. In the course of conducting this research, we developed a new design for the construction of Faraday isolators. This new Faraday isolator shows performance superior to that of conventional designs.				
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Final Report

Robert W. Boyd – Principal Investigator
George L. Fisher – Graduate Research Assistant

Our research falls into two main categories. The results in each area are reported in turn.

Enhancement of Third-order Optical Nonlinearities via Composite Formation

As a test of recent theory, we have constructed layered composites and measured their linear and nonlinear optical properties. We have measured a thirty-five percent enhancement in the third-order nonlinear optical properties of our samples.

Background and Theory The need for materials possessing large optical nonlinear coefficients for use in nonlinear optics and in photonics is great. Much nonlinear optical materials research is devoted to the development new organic polymers with ever higher values of the third-order susceptibility $\chi^{(3)}$. Our approach is to find a method to increase the nonlinear response of a broad class of existing materials. Recently, several groups have reported on the potential use of composite materials as a way to increase $\chi^{(3)}$. The method we have chosen, proposed by Robert Boyd and John Sipe, involves forming a composite made of alternating layers of two different materials with layer thicknesses much smaller than an optical wavelength. If the linear refractive indices of these two components are different, the electric field strength of an optical field polarized perpendicular to the plane of the layers will be nonuniformly distributed between the two constituent materials. From fundamental electro-magnetic theory, the field strength will exceed the spatially averaged field strength in the material with the smaller linear refractive index. If this is the more nonlinear material of the two then the effective $\chi^{(3)}$ value of the composite can exceed the $\chi^{(3)}$ value of a sample of pure nonlinear material with thickness equal to the entire composite thickness. As an example, if the refractive indices differ by a factor of two, the value of $\chi^{(3)}$ can be enhanced by a factor of ten. This tenfold increase occurs when the total thickness of the nonlinear layers in the composite is only approximately 30% of the total thickness, so it is possible to get a higher signal while using less of the optically active material.

This maximum enhancement occurs when the incident light propagates through the material as a TM guided wave. Smaller enhancements are predicted for light traveling through the sample at some oblique angle. While not the end goal of our work, these smaller enhancements provide a means to test our theory. The angular dependence of the nonlinear optical response can be used to quantitatively determine the degree to which the $\chi^{(3)}$ susceptibility is enhanced. We have performed a thorough experimental study of this angular dependence. We find that the angular dependence is in excellent agreement with our theoretical predictions, and quite different from predictions based on theory which does not contain enhancement due to local field effects.

Composites We have formed several composite samples. Each consists of alternating layers of titanium dioxide and the nonlinear optical polymer PBZT (poly(paraphenylene benzobisthiazole)). In our samples titanium dioxide is used as the high-index component having a nonlinear response orders of magnitude smaller than PBZT. PBZT was chosen for its high nonlinearity, good processability, and for its availability. The samples are formed by spin casting onto glass substrates. The titanium dioxide is spin

Third-order nonlinear susceptibility of a family of conjugated organic polymers.

In this project we measured both the sign and the magnitude of the third-order nonlinear susceptibility for a family of five substituted conjugated organic polymers.

Polymers Our sample set consisted of poly (p-phenylene benzobisthiazole) PBZT, poly (biphenylene benzobisthiazole) PBBZT, poly (2,6-naphthalene benzobisthiazole) 2,6-PNBT, poly (1,4-naphthalene benzobisthiazole) 1,4-PNBT, and poly (benzimidazole-1,4-phenylenebisvinylene) PBIPV. The polymers were synthesized by Samson Jenekhe's group in the Department of Chemical Engineering at the University of Rochester.

Laser System $\chi^{(3)}$ was measured for the polymers using the z-scan technique. A Q-switched, mode locked, Quantel Nd:YAG laser supplied 30 ps pulses at 10 Hz. The pulse with the largest energy is electro-optically selected from each pulse train and amplified. In order to avoid two photon absorption of the 1.06 μm wavelength light in the samples, the fundamental output of the laser was Stoke's shifted to 1.9 μm in a Raman cell containing hydrogen at 35 atm. In the Raman cell some of the incoming photons excite hydrogen molecules to higher vibrational states. To conserve energy the emitted photons have lower energy, hence the 1.9 μm wavelength output light.

Results The susceptibilities we measured are tabulated in the table below. Absolute $\chi^{(3)}$ values are in agreement with the $\chi^{(3)}$ values in the table which are relative to the $\chi^{(3)}$ of CS_2 . The values are very large in magnitude and negative in sign. We have written a preliminary version of a journal publication on this project.

Material	$\chi^{(3)} \times 10^{-9}$ esu
1,4 PNBT	-2.8 ± 0.3
PBZT	-2.7 ± 0.3
2,6 PNBT	-2.7 ± 0.3
PBBZT	-1.9 ± 0.2
PBIPV	-1.6 ± 0.2

Improved Faraday Rotator

We have modeled, built, and tested a Faraday rotator for wavelengths of 1.06 μm and shorter. Our Faraday rotator employs two novel features which were discovered in our laboratory, small gaps between magnets and a nonsymmetrically placed rotator rod.

Computational Using standard theory, we modeled the strength of the magnetic field at all points away from the pole face of a magnet. We used this information to calculate the rotation of the plane of polarization for polarized light propagating through rotator rods which, in turn, are influenced by magnetic fields. After varying several parameters we discovered that separating the magnets can in some cases increase the rotation and also increase the uniformity of rotation across the diameter of the rod. We also confirmed our intuition that a nonsymmetrically placed rotator rod is ideal for a tunable Faraday rotator.

Construction We designed and machined our own holder to keep the one Tesla magnets together. The magnets are oriented to repel each other and must be brought together. The one cm holes in each magnet must also line up in order to have the maximal clear aperture and optimal functioning of the device. Since we wanted to test the rotator in a variety of configurations we opted for a modular design.

cast from a sol gel precursor and is cured for 24 hours at 200 C to yield a material with a refractive index of approximately 2.2. The PBZT is cured for 24 hours under vacuum at 70 C after it is spin cast. PBZT has a linear refractive index of 1.7 and a $\chi^{(3)}$ of 5×10^{-11} esu. Layer thicknesses of 50 nm for the titanium dioxide and 40 nm for the PBZT are used. The samples used in the measurements described below contain five layers of each material.

Experiment The nonlinear optical response of these materials is measured as a function of the angle of incidence using the z-scan method. Measurements are performed at a wavelength of 1.9 μm to avoid two-photon absorption in the polymer portions of the samples. This wavelength is obtained by Raman shifting the 1.06 μm fundamental output of a Nd:YAG in a hydrogen cell at 35 atm. The laser is configured with a 0.3 mm etalon to produce 30 ps pulses. The use of short pulses minimizes thermal effects in the nonlinear optical response as well as increases the intensity of the pulse. The prism is used in conjunction with filters to separate the 1.9 μm light from any other wavelength light which either makes it through the Raman cell unshifted, or is created by the cell.

Results Results of our measurements are shown in Fig. 1. Here the nonlinear phase shift is plotted as a function of angle of incidence for both TM (p-polarized) and TE (s-polarized) light. The solid curves are theoretical. They are completely determined by our measured values of the linear properties of the materials in the composite. There are no free parameters in our theory. The dashed curves are what we would expect without enhancement. Since there is no enhancement in either theory for TE light, the two TE curves are collinear. Note that the nonlinear phase shift first increases and then decreases with angle as the Fresnel transmission coefficient drops to zero at grazing incidence for the TM wave theoretical curve which includes enhancement. The excellent agreement between theory and experiment indicates that the local field effect theory is correct and that the formation of layered composites is a valid method of enhancing optical nonlinearities. Portions of this work have been presented at two conferences in the form of full length presentations and at two conferences in the form of poster presentations. A final manuscript has been prepared for submission to *Physical Review Letters*.

Testing We tested both the rotation and the uniformity of rotation for a seven-magnet three-rotator-rod Faraday rotator and for a three-magnet one-rotator-rod Faraday rotator. Using the seven magnet Faraday rotator we varied the magnet-magnet separation from zero to ten mm and measured the rotation and the uniformity at each position. The data agreed with our computer modeling. The introduction of a gap between magnets can increase rotation and uniformity. Using the three magnet Faraday rotator we moved first a 25 mm rotator rod from centered in the middle magnet to centered in an end magnet and measured rotation and uniformity and next did the same for a 17 mm long rotator rod. Here again we had very good agreement between our data and our computer models. A rod shorter than the central magnet gives excellent uniformity for a fixed wavelength rotator, but for a tunable rotator the best Faraday rotator has an offset rod equal in length to that of the central magnet.

Results Since we came up with two potentially useful results for the construction of Faraday rotators we will publish our findings. We have a finished manuscript ready for submission to the *Journal of Modern Optics*. Our two main improvements are the introduction of a separation between magnets and the displacement of the rotator rod.